Controlled Stark shifts in Er^{3+}-doped crystalline and amorphous waveguides for quantum state storage

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We present measurements of the linear Stark effect on the ^{4}I$_{15/2} \rightarrow$ ^{4}I$_{13/2}$ transition in an Er^{3+}-doped proton-exchanged LiNbO$_3$ crystalline waveguide and an Er^{3+}-doped silicate fiber. The measurements were made using spectral hole burning techniques at temperatures below 4 K. We measured an effective Stark coefficient $(\Delta\mu_e\chi)/(h) = 25 \pm 1$kHz/Vcm^{-1} in the crystalline waveguide and $(\overline{\Delta\mu_e}\chi)/(h) = 15 \pm 1$kHz/Vcm^{-1} in the silicate fiber. These results confirm the potential of Erbium doped waveguides for quantum state storage based on controlled reversible inhomogeneous broadening.

Because of their narrow homogenous line widths and corresponding long optical coherence times at low temperatures, rare-earth doped crystals (RE doped crystals) have been the subject of much investigation for photon-echo based optical data storage and data processing [1]. Recently, there has been additional interest in RE doped crystals, spurred by proposals to extend their use to applications in the fast developing field of quantum communication and computation [2]. In particular, RE doped crystals are a promising material for the realization of a reversible transfer of quantum states between photons and atoms. Such a quantum memory represents a basic building block for the so-called quantum repeater [3], which allows the extension of quantum communications schemes such as quantum cryptography [4] to unlimited distances.

A original proposal for a quantum, as well as classical, memory in solid state material is based on controlled reversible inhomogeneous broadening (CRIB) of a narrow, spectrally isolated absorption line [5]. This protocol requires an atomic ensemble with a large optical depth, a long optical coherence time, and the ability to inhomogeneously broaden a single absorption line in the ensemble in a controlled and reversible way.

Erbium doped waveguides are promising materials for the realization of CRIB, as interaction lengths ranging from many cm (in the case of crystalline waveguides), to hundreds of meters or more (in the case of amorphous optical fibers) can easily be achieved, allowing for large absorption even at low doping concentrations. The 1.5 μm, ^{4}I$_{15/2} \rightarrow$ ^{4}I$_{13/2}$, transition in Er^{3+} is well matched to standard telecommunication fiber, which would allow simple interfacing of such a memory in a fiber communication network. In addition, recent investigations show that coherence times on the scale of μs can be obtained in Er^{3+} doped silicate fibers [6, 7] , and on the order of

ms in Er^{3+} doped crystals [8]. In this article we investigate the linear Stark effect for the realization of CRIB in two different Erbium doped waveguides, a crystalline LiNbO$_3$ waveguide and a silicate optical fiber. Our results confirm the potential of Erbium doped waveguides for CRIB-based data storage.

In the presence of an external DC electric field, the energy levels of an atom with a permanent dipole moment are shifted; this phenomenon is known as the linear DC Stark effect. If the dipole moments are different for different electronic levels this shift leads to a shift in the associated optical transition frequency. The linear DC Stark effect can be observed in RE doped solids, where the dipole moment in the ion is induced by local electric fields. It has been measured for the ^{4}I$_{15/2} \rightarrow$ ^{2}H$_{11/2}$ transition in Er^{3+}:LiNbO$_3$ [9] and Er^{3+},Mg^{2+}:LiNbO$_3$ [10] crystals, using a differential technique [11]. In addition the effect has been measured in RE doped bulk crystals such as Eu^{3+}:YAlO$_3$ [12, 13], Pr^{3+}:YSiO$_5$ [14], Pr^{3+}:YAlO$_3$ [15] and Pr^{3+}:LaF$_3$ [16] as well as in Sm^{2+} doped borate bulk glass [17], an amorphous material. In order to achieve high sensitivity in the latter measurements, spectral hole burning techniques [18] are used to isolate the effect of an electric field on a single homogenous line within an inhomogeneously broadened ensemble of atoms. To the best of our knowledge no measurements have been reported on RE doped waveguides, neither crystalline nor amorphous.

We consider the linear Stark effect where the transition frequency, ω, of an ion is shifted by the electric field according to the formula,

$$\Delta\omega = \frac{\Delta\mu_e\chi E}{\hbar}\cos\theta. \qquad (1)$$

Here $\Delta\mu_e$ is the difference between the permanent elec-

tric dipole moments of the two states connected by the transition, E is the applied DC electric field, $\chi = (\epsilon+2)/3$ is the Lorentz correction factor, ϵ is the dielectric constant of the sample, and θ is the angle between the vectors $\Delta\vec{\mu}_e$ and $\vec{E}$.

In a crystal, because of the ordered structure of the crystallin lattice, the dipole moments are aligned along a set of one or more well defined directions. The symmetry of the crystal along with the site symmetry at the rare-earth-ion position determines the number of directions. All ions with $\Delta\vec{\mu}_e$ aligned along the same direction experience the same shift in their resonance frequency with an applied electric field and thus the application of a DC electric field leads to a shift or splitting of the spectral hole, depending on the number of possible directions. The projection of $\Delta\mu_e$ between the two states, along the direction of the applied electric field, can be determined by measuring the shift of the hole as a function of the electric field, as given by equation 1.

In an amorphous material, such as an optical fiber, the dipole moments are randomly orientated, such that the projection of $\Delta\vec{\mu}_e$ along the direction of the applied electric field varies continuously for different ions in the solid. Therefore, rather than a shift or splitting of the spectral hole, one observes a broadening as the absorption frequencies shift by different amounts, in different directions. Assuming a Lorentzian spectral hole, one can derive the shape of the hole for a given applied electric field. Here we make the following simplifying assumptions as proposed by Bogner et al. [19]. First, the dipole moments are randomly orientated in the solid. Second, there is a distribution of magnitudes of $\Delta\mu_e$ which can be described by a Maxwell Distribution:

$$g(\Delta\mu_e) = \frac{4}{\pi^{1/2}(\overline{\Delta\mu_e})^3}(\Delta\mu_e)^2 \exp\left[-\left(\frac{\Delta\mu_e}{\overline{\Delta\mu_e}}\right)^2\right]. \quad (2)$$

Here $\Delta\mu_e$ is the difference in dipole moment between the two states, and $\overline{\Delta\mu_e}$ is the most likely value of this difference. Note that the width of the distribution and the value of $\overline{\Delta\mu_e}$ are not independent.

With these assumptions the shape of the spectral hole, $h(x)$, under application of an electric field, is given by [20]

$$h(x) = \frac{2}{\pi^{3/2}\overline{f}^3}\left\{\int_0^{\sqrt{1+x^2}} f \exp\left[-\left(\frac{f}{\overline{f}}\right)^2\right] * \right.$$
$$\arctan\left(\frac{2f}{1-f^2+x^2}\right) df + \quad (3)$$
$$\int_{\sqrt{1+x^2}}^{\infty} f \exp\left[-\left(\frac{f}{\overline{f}}\right)^2\right] *$$
$$\left.\left[\pi + \arctan\left(\frac{2f}{1-f^2+x^2}\right)\right] df\right\}$$

with

$$\overline{f} = \frac{\overline{\Delta\mu_e}\chi E}{\hbar\gamma} \qquad x = \frac{(\omega - \omega_C)}{\gamma}. \quad (4)$$

Here ω is the laser frequency, ω_C is the central transition frequency, γ is the full width at half maximum (FWHM) of the spectral hole without applied electric field, and the other variables are defined as in equation 1.

In our investigation of the Stark effect in Er^{3+} doped waveguides we used an monochromatic laser to excite Er^{3+} ions from the $^4I_{15/2}$ ground state to the $^4I_{13/2}$ excited state, thereby creating a spectral hole at the frequency of the laser. We then decreased the laser intensity and scanned the frequency around the initial burning frequency three times while measuring the transmitted light as a function of time. Together with an independent time-to-frequency calibration of the laser scan, this yields three spectral hole profiles. During the first scan there was no applied electric field, for the second scan we applied an electric field across the sample, as described below. Finally, for the third scan, we switched off the electric field to demonstrate the reversibility of the Stark induced line shift.

The first sample, a single domain, z-cut, proton-exchanged $LiNbO_3$ waveguide (dimensions 1 mm thick, 6 mm long, 6 mm wide) is doped with Er^{3+} (0.5 at %), and MgO (5 mol %). We mounted the waveguide between two metal electrodes spaced by 1 mm and cooled it to 3.8 K using a pulse tube cooler (VeriCold Technologies). Light, linearly polarized along the crystal C_3 symmetry axis, was injected and collected with standard optical fibers mounted on two xyz positioners (Attocube Systems) and aligned with the input and output of the waveguide. The C_3 symmetry axis was oriented parallel to the applied electric field and perpendicular to the light propagation direction. In addition we applied a magnetic field of approximately 0.7 T parallel to the C_3 symmetry axis to reduce the width of the spectral hole [21]. We performed hole burning experiments as described above, with applied voltages ranging from -200 to 200 V, using a scannable cw external cavity diode laser (Nettest, Tunics Plus) at a wavelength of 1531.00 nm. To measure the spectral hole we scanned the laser over 1.2 GHz in 500 μs and measured the transmitted light with a photodiode (NewFocus, mod. 2011).

The frequency shift of the spectral hole, relative to the zero field position, as a function of electric field is plotted in figure 1. The line is a linear fit to the data which passes through the point (0,0). The dipole moment difference can be calculated from the slope of the line according to equation 1. We find an effective dipole moment difference $(\Delta\mu_e\chi)/(h) = 25 \pm 1$kHz/Vcm^{-1}. The inset graph in figure 1 shows the position of the spectral hole measured after switching off the electric field for each applied field;
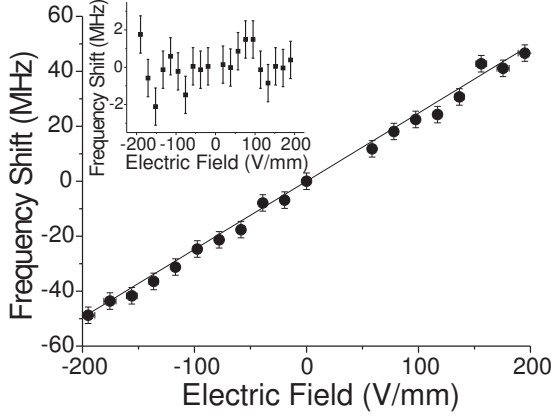
FIG. 1: The shift of the spectral hole central frequency as a function of electric field in the Erbium doped $LiNbO_3$ waveguide. The line is a linear fit to the data, which passes through the point (0,0), and gives an effective dipole moment difference of $(\Delta\mu_e\chi)/(h) = 25 \pm 1\text{kHz/Vcm}^{-1}$. The inset graph shows the frequency shift of the hole after the applied field is turned off.

the spectral hole returns to the initial zero field position. This, plus the opposite directional shifts that were observed with positive and negative voltages, confirms the reversibility of the Stark effect.

The observed single-frequency shift of the absorption line, under application of an electric field in the direction of the C_3 symmetry axis, reflects the lack of inversion symmetry in the $LiNbO_3$ crystal. This property allows for a single dipole moment direction, in contrast to observations of a "pseudo Stark splitting" [23] in centrosymmetric crystals, where the inversion symmetry results in dipole moments pointing in opposite directions.

The second sample, a SiO_2 fiber (Ino, ER 407 Fiber) is doped as follows: Er 0.07 (at %), Al 2.65 (at %), Ge (3.62 at %). The fiber (length 65 cm) was coiled with a diameter of 4 cm and sandwiched between two round metal electrodes spaced by 0.3 mm. We placed the sample in a $^3\text{He}/^4\text{He}$ dilution refrigerator and cooled it to 50 mK. As the spectral hole width is sufficiently small at this temperature, application of a magnetic field was not necessary. We performed spectral hole burning experiments as described above, with applied voltages ranging from 0 to 120 V, using a cw external cavity diode laser (Toptica, DL 100) at a wavelength of 1531 nm. To measure the spectral hole we scanned the laser over 400 MHz in 800 μs. The transmission was measured with a photodiode (NewFocus, mod. 2011).

The spectral hole for each applied electric field is plotted in figure 2a. The points are the measured data and the lines are fits using equation 4. We find a close agreement between the predicted and observed hole shapes. The values of f obtained from the fits for each applied electric field are plotted in figure 2b. The line is a linear fit to the data which passes through (0,0). The dipole
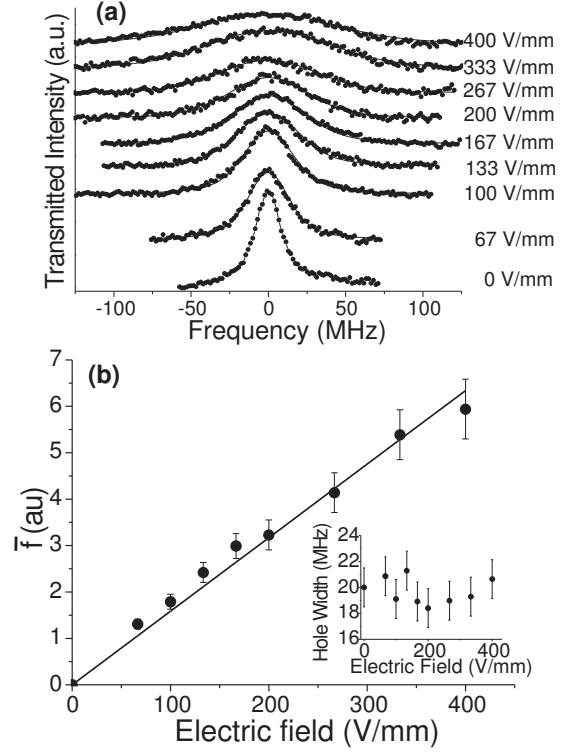

FIG. 2: Spectral holes in Er^{3+} doped silicate telecom fiber, as a function of applied electric field. Figure (a) shows the spectral holes (transmitted intensity as a function of frequency) with different applied electric fields, the holes are shifted vertically for clarity. The points are measured data and the lines are a fit to equation 3. Figure (b) is a plot of the fitted $\overline{f}$ values from figure (a). The line is a linear fit to the data, which passes through the point (0,0), and gives an effective dipole moment difference $(\overline{\Delta\mu_e\chi})/(h) = 15 \pm 1\text{kHz/Vcm}^{-1}$. The inset graph in figure (b) shows the width of the spectral hole (FWHM) measured after the applied field is turned off.

moment difference can be calculated from the slope of the line according to equation 4. We find an effective dipole moment difference $(\overline{\Delta\mu_e\chi})/(h) = 15 \pm 1\text{kHz/Vcm}^{-1}$. The inset graph in figure 2b shows the width of the spectral hole (FWHM) measured after switching off the electric field for each applied field. The spectral hole width returns to the initial zero field value, which confirms that the broadening of the spectral hole can be removed.

Let us now discuss our results in view of the controlled reversible inhomogeneous broadening required for the quantum memory proposal [5]. The broadening must be controllable in such a way that, starting with an ensemble of ions that absorb at ω_0, the transition frequency of the ion can be shifted to $\omega_0 + \omega_b$ and then, at a later time, shifted again to $\omega_0 - \omega_b$, where ω_b varies continuously over the necessary bandwidth. For the storage of light pulses with a duration of 10ns, for example, the required frequency bandwidth is on the order of 100 MHz. In the crystal this broadening could be achieved by applying a position dependent electric field along the crystal,

ranging from $+2$ kV cm^{-1} to -2 kV cm^{-1}. In the fiber, the application of a spatially homogeneous electric field of approximately 3.5 kV cm^{-1} is sufficient as it directly leads to a broadening of the spectral hole. In both the fiber and the crystal, switching the polarity of the applied field should reverse the broadening in the required way.

In conclusion, we have presented measurements of the linear DC Stark effect in an Er^{3+}-doped proton-exchanged LiNbO$_3$ waveguide, and an Er^{3+} doped silicate telecom fiber. We found effective dipole moment differences of $(\Delta\mu_e\chi)/(h) = 25 \pm 1$kHz/Vcm^{-1} and $(\overline{\Delta\mu_e}\chi)/(h) = 15 \pm 1$kHz/Vcm^{-1}, respectively. These measurements demonstrate the suitability of Er^{3+} doped waveguides for solid state quantum memory protocols based on controlled reversible inhomogeneous broadening.

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